



# Phase-field simulations of microstructure evolution during physical vapor deposition of single-phase thin films



James A. Stewart<sup>a</sup>, Douglas E. Spearot<sup>b,\*</sup>

<sup>a</sup> Institute for Nanoscience & Engineering, University of Arkansas, Fayetteville, AR 72701, USA

<sup>b</sup> Department of Mechanical & Aerospace Engineering, University of Florida, Gainesville, FL 32611, USA

## ARTICLE INFO

### Article history:

Received 18 January 2017

Accepted 19 January 2017

### Keywords:

Phase-field modeling  
Physical vapor deposition  
Polycrystalline thin films  
Microstructure evolution

## ABSTRACT

Physical vapor deposition (PVD) simulations are performed using a phase-field model to study microstructure evolution in single-phase polycrystalline materials as a function of deposition conditions. Specifically, this work focuses on the influence of (i) polycrystalline substrate microstructure with low-angle and high-angle grain boundaries (GBs), (ii) incident vapor flux rate, and (iii) model parameters controlling grain evolution dynamics. Simulation results show that low-angle and high-angle GBs in the substrate promote the formation of low-angle and high-angle GBs within the deposited thin film, in qualitative agreement with experimental reports. Changing the vapor flux rate has a significant influence on the deposited microstructure. A relatively low flux rate is found to provide a thin film with no subsurface porosity, smoothed surface roughness, and a mixture of grain sizes. With an increased vapor flux rate, uniformly distributed surface columnar features are observed with subsurface porosity. The GBs migrate during growth to align with these surface features. At the largest flux rate studied, a dendritic style microstructure is formed where the GBs tend to align with and grow along the regions of low phase density within the thin film. Finally, a parametric study is performed on the phase-field model grain evolution parameters to elucidate their role on grain boundary thickness, internal grain variations and grain rotation during simulated PVD.

© 2017 Elsevier B.V. All rights reserved.

## 1. Introduction

Thin films grown by physical or chemical vapor deposition methods usually have surface and subsurface features (columnar structures, grains, grain boundaries (GBs), etc.) with sizes in the nanometer to micrometer range. The surface morphology and underlying microstructure of a thin film are primarily determined by the specific deposition conditions, method, and materials used. Furthermore, the details of the surface morphology and subsurface microstructure, such as crystal structure and orientation, grain size, and chemical composition, greatly influence the mechanical, electrical, and chemical properties of the thin film. These properties dictate the possible applications, e.g., optoelectronic and microelectronic devices, nanoelectromechanical systems, or optical and protective coatings [1–8]. As such, to study the fundamental properties of thin films, it is necessary to consider the formation and evolution of surface and subsurface features for a physically

relevant system (e.g., grains and GBs in polycrystalline materials) under given vapor deposition conditions.

The focus of this modeling and simulation work is on the physical vapor deposition (PVD) of single-phase polycrystalline materials. PVD is selected because, during transport and thin film growth, the composition of the depositing target material is conserved and physical processes dominate over chemical reactions [4,6]. Therefore, chemical reactions and composition changes can be neglected in the development of a computational model provided the material under consideration is composed of a single element or a compound where all of the phases have the same stoichiometry. As such, it is necessary to include evolutionary dynamics that are fundamental to the PVD growth process, for example: arbitrary surface formation, surface tension and diffusion, and nonlocal shadowing effects [4–6]. By utilizing modeling and simulation techniques with proper descriptions of the underlying physics for PVD and polycrystalline evolution, the goal is to elucidate influences of PVD process parameters on grain and GB evolution during thin film growth.

Recently, Stewart and Spearot [9] developed a phase-field model for simulating microstructure development during PVD of a single-phase polycrystalline material. The phase-field method is

\* Corresponding author.

E-mail address: [dspearot@ufl.edu](mailto:dspearot@ufl.edu) (D.E. Spearot).

a popular simulation technique that has been successfully utilized to study complex physical processes without explicit interfacial tracking, e.g., solidification of an undercooled liquid, solid-state phase transformations, grain growth, pattern formation in alloys, and more recently polycrystalline vapor deposition [9–14]. In this technique, field variables are assumed to exist, which describe physical quantities of the system at every location within the model, such as structural order, composition, or orientation. The free energy of the system is constructed as a functional of these field variables, their spatial gradients, and other thermodynamic or applied forces influencing energetics [10–14]. Evolution of the system is then driven by minimization of the free energy, which is governed by Allen-Cahn or Cahn-Hilliard dynamics for non-conserved and conserved phenomena, respectively [10–12].

In prior work, Stewart and Spearot [9] focused on the mathematical development of a free energy functional, numerical implementation, and application of the model to study the role of the substrate grain size on microstructure development during PVD growth of a thin film. In the current work, the PVD phase-field model developed in [9] is applied to simulate isothermal PVD of a generic single-phase polycrystalline metal to qualitatively study the influence of several PVD process attributes on grain and GB evolution: (i) the role of substrate microstructure using substrates with only low-angle GB misorientations and both low-angle and high-angle GB misorientations, (ii) the role of varying incident vapor flux rate, and (iii) the influence of grain and GB kinetics on the growth of the thin film microstructure. Thus, this work explores several important attributes of the PVD process not previously considered in [9].

## 2. Simulation methodology

In this section, a brief overview of the PVD phase-field model developed by Stewart and Spearot [9] is given. This model was shown to provide a simulation framework for simultaneous thin film growth, surface feature formation, grain growth, and GB migration during PVD. The underlying physics incorporated into this model have been shown to capture aspects relevant to PVD and polycrystalline evolution, including arbitrary surface formation, surface tension and diffusion, shadowing effects, and grain coarsening and impingement; in depth discussions of these aspects can be found in [15,16].

### 2.1. Phase-field PVD model

To model PVD and polycrystalline evolution within the phase-field framework, three field variables are introduced:  $f(\mathbf{r}, t)$ ,  $\theta(\mathbf{r}, t)$ , and  $\mathbf{g}(\mathbf{r}, t)$ . The first field variable,  $f(\mathbf{r}, t)$ , describes the evolution of the growing thin film solid where  $f(\mathbf{r}, t) \approx 1$  describes the solid phase,  $f(\mathbf{r}, t) \approx -1$  describes the vapor phase, and  $f(\mathbf{r}, t) \approx 0$  defines the solid-vapor interface [15]. The second field variable,  $\theta(\mathbf{r}, t)$ , describes the local grain orientation as measured from a defined global axis (e.g., the positive  $x$ -axis). These two field variables and their gradients are used to construct the free energy functional for describing PVD of a single-phase polycrystalline material, as shown in Eq. (1).

$$F = \int \left( -\frac{1}{2}f^2 + \frac{1}{4}f^4 + a(\nabla f)^2 + sf_d^2|\nabla\theta| + \frac{\varepsilon^2}{2}f_d^2|\nabla\theta|^2 \right) d\Omega \quad (1)$$

The first two terms in Eq. (1) provide a double-well potential describing the bulk solid and vapor phases. The third term provides an energy penalty for the presence of the solid-vapor interface where  $a$  is the interfacial gradient coefficient, which is related to surface tension. The fourth and fifth terms account for energy contributions related to grain misorientation. Warren et al. proved

that the linear term in  $|\nabla\theta|$  is the only solution that provides a finite width GB (introduces a cusp in the free energy) and the quadratic term in  $|\nabla\theta|$  allows GB motion [16–18]. Furthermore, constructing the free energy functional using grain misorientation maintains the requirement that bulk energy contributions are reference frame invariant. The grain gradient coefficients are taken to be products of  $s$ ,  $\varepsilon$ , and  $f_d$ . The parameters  $s$  and  $\varepsilon$  can be related to the latent heat of fusion and a characteristic GB thickness (cf. [16]), while the local solid density  $f_d(\mathbf{r}, t) = (f(\mathbf{r}, t) + 1)/2$  is introduced so that GB energy contributions are removed within the vapor region. Finally, the third field variable,  $\mathbf{g}(\mathbf{r}, t) \geq 0$ , which doesn't contribute to the free energy, describes the local density of the incident vapor, where  $\mathbf{g}(\mathbf{r}, t) \approx 0$  specifies the presence of no vapor.

Using the free energy functional in Eq. (1), the equations of motion for PVD of a single-phase polycrystalline thin film can be determined. First, it is assumed that the growth of the solid phase,  $f(\mathbf{r}, t)$ , occurs at the expense of the incident vapor phase,  $\mathbf{g}(\mathbf{r}, t)$ , with appropriate influences from the polycrystalline microstructure. This is expressed mathematically in Eqs. (2) and (3) as was proposed by Koblinski et al. [15].

$$\frac{\partial f}{\partial t} = \nabla^2 \frac{\delta F}{\delta f} + B(\nabla f)^2 \mathbf{g} + C\sqrt{(\nabla f)^2} \eta \quad (2)$$

$$\frac{\partial \mathbf{g}}{\partial t} = \nabla[D\nabla \mathbf{g} - \mathbf{A}\mathbf{g}] - B(\nabla f)^2 \mathbf{g} \quad (3)$$

In Eq. (2), the first term provides Cahn-Hilliard dynamics, which allows for arbitrary surface formation while simultaneously capturing surface diffusion. The second term is the source term that couples Eq. (2) with Eq. (3) and allows for growth of the thin film at the expense of the vapor phase, where the coefficient,  $B$ , controls the growth rate. The last term provides surface fluctuations through a Gaussian noise distribution,  $\eta(\mathbf{r}, t)$ , where the parameter,  $C$ , controls the noise amplitude. Next, Eq. (3) describes transport of the incident vapor through the diffusion equation modified for the presence of an external force,  $\mathbf{A}$ . Here,  $D$  is the diffusion coefficient and  $\mathbf{A}$  provides direction and strength to the incident vapor. The second term is the negative of the second term in Eq. (2) and acts as a sink that removes vapor that has been converted to solid.

Finally, Eq. (1) is used to determine equations of motion describing subsurface grain and GB evolution through Allen-Cahn dynamics, which gives rise to Eq. (4), where  $\tau_\theta$  controls the overall rate of grain evolution and  $P(\varepsilon|\nabla\theta|)$  is a function that modulates the rate of grain evolution in grain interiors and near GBs.

$$P(\varepsilon|\nabla\theta|)\tau_\theta f_d^2 \frac{\partial \theta}{\partial t} = \nabla \cdot \left[ f_d^2 \left( \frac{s}{|\nabla\theta|} + \varepsilon^2 \right) \nabla\theta \right] \quad (4)$$

Eq. (4) allows grain rotations to take place, causing grain misorientations to change. Therefore, a crystal with a given rotational symmetry,  $N$ , needs to strictly be considered when calculating energetic contributions. This is enforced by mapping all  $\theta(\mathbf{r}, t)$  values to the domain  $-\pi/N < \theta(\mathbf{r}, t) \leq \pi/N$  [16].

It is worthwhile to reiterate several aspects of the grain evolution model. In the formalism developed by Warren et al. [16], only the term linear in  $|\nabla\theta|$  is required to consider a static GB and this formulation captures classical (Read-Shockley) relationships between GB energy and misorientation, enabling differentiation between low-angle and high-angle behavior. Kobayashi et al. showed [20] that at static equilibrium, the solution for the orientation field profile at the GB is a step function (discontinuity). However, when considering dynamic GB behavior (motion and grain rotation), the term quadratic in  $|\nabla\theta|$  in Eq. (1) must also be included [20,21]. This term smooths the cusp in the phase field parameter at the interface and promotes a sigmoidal shape for the orientation transition between grains, leading to a diffuse interface whose thickness depends on the magnitude of the misori-

Download English Version:

<https://daneshyari.com/en/article/5453232>

Download Persian Version:

<https://daneshyari.com/article/5453232>

[Daneshyari.com](https://daneshyari.com)